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Title: **THERMAL RESPONSE BASED ITEM
IDENTIFICATION**

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Submitted to: **42nd Annual INMM Meeting
Indian Wells, CA USA
July 15-19, 2001
(FULL PAPER)**

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THERMAL RESPONSE BASED ITEM IDENTIFICATION

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*Presented at the
Institute of Nuclear Material Management
42nd Annual Meeting
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ABSTRACT

One of the most difficult problems in NDA of nuclear materials is identifying the chemical form of the nuclear material and the surrounding matrix. Recent work analyzing the calorimeter response of sources embedded in a variety of matrices has led to a possible solution to this problem. The wide range of thermal time constants exhibited by typical matrix materials lends itself to permitting the differentiation between materials, based on time constants extracted from the measured response. Potential applications include simple item identification, item fingerprinting as part of shipper-receiver measurements, and distinguishing between Pu metal and Pu oxide as required under certain proposed attribute measurements. The results of applying this technique to a variety of items will be presented and discussed.

INTRODUCTION

Calorimetry has a long history as a nondestructive assay (NDA) tool for measuring quantities of nuclear material. In this area, it has been used exclusively to measure the power output of an item. The power is combined with isotopic information to establish the mass of nuclear material in a particular item. This technique has been very successful and currently calorimetry combined with isotopics is the highest-precision and lowest-bias NDA method for determining the mass of nuclear material.

While it has been clear that more information than the power output is available from the calorimeter response, it has been difficult to extract that information. In general, if more detailed information has been required, it has been possible to do destructive analysis (DA) on an item to extract material properties. Over the last few years, there has been a growing need to extract material properties from items containing nuclear materials using NDA methods alone. One example is the need under proposed verification agreements to determine whether an item contains plutonium metal or plutonium oxide.[1]

Recently, a series of careful measurements of various items has been carried out and analyzed. The results of the analysis demonstrate that calorimetry could be used to differentiate between different matrices independent of the specific item output power. The details of the measurements and the analysis will be presented along with conclusions, potential applications, and future work.

EXPERIMENTAL METHOD

A series of measurements was taken in a calorimeter with a measurement chamber size of 12.7 cm in diameter by 25.4 cm in height. The calorimeter is located in the Safeguards Science and Security Calorimetry Laboratory. The calorimeter can was filled with different matrices and preequilibrated to 24 degrees Celsius, one degree below the

temperature of the HEU calorimeter bath. This preequilibration was accomplished by placing the calorimeter can in another calorimeter equipped with a high precision bath controller, accurate to 0.001 degrees Celsius. The preequilibration controller was set to 24 degrees Celsius. The HEU calorimeter bath controller was set to 25 degrees Celsius. Preequilibration was performed to ensure that all matrices were at the same temperature when they were inserted into the calorimeter. The can was left in the preequilibrator for 12 to 36 hours, depending on the thermal properties of the matrix.

Matrices investigated included air, aluminum (as crumpled foil or a combination of crumpled foil and solid bars), copper shot, lead shot, steel shot, polyethylene beads, salt tablets, and sand.

For each matrix, calorimetry measurements were taken with and without a ^{238}Pu heat source (0.8 Watts) in the calorimeter. The mass of matrix material was determined for each run by weighing the can and contents on a Pennsylvania Model 7500 digital scale. The empty can was also weighed, which allowed us to calculate the mass of matrix material present for each measurement. The volume of the calorimeter can was approximately 3 liters. The measurements were ended when careful visual inspection determined that equilibrium had been reached and maintained for at least four hours.

Data acquisition was managed by MultiCal.[2] Data points were read and recorded every two seconds. The program monitored bridge potential, bridge current, and bath temperature. The data of interest is the bridge potential data. The bridge current and bath temperature were monitored for diagnostic purposes. Data from each run was extracted from the MultiCal database and transferred to another computer for the data analysis.

DATA ANALYSIS

The data analysis method was a direct outgrowth of ongoing work aimed at developing better equilibrium prediction and detection algorithms. This work has been discussed in detail elsewhere. [3] Briefly, the response of a calorimeter can be modeled by a function of the form,

$$f(t) = A + \sum_i B_i e^{-t/C_i}, \quad (1)$$

where $f(t)$ is the calorimeter response, either bridge potential or servo power, as a function of time, t . While an algorithm has been developed to fit the above equation with an arbitrary number of exponential terms in an automated way, a different approach was used for the data presented here. Since only a small number of assays were to be analyzed and a method for estimating uncertainties was required, a similar procedure using Microsoft ExcelTM was implemented that included an uncertainty analysis.

The raw data was obtained at measurement intervals of about 2 seconds. In order to reduce the data to a manageable number of points, a subset of the data was extracted, composed of measurements made 20 seconds apart. The assumption was made that the large number of points that remained (typically 2000-5000) was sufficient to characterize the overall response. This was not done for the measurement made with copper shot without a source. Those data were rebinned into 120-second intervals shortly after

acquisition, and the original data could not be recovered. Therefore, the binned data was used without resampling.

Each of the assays was loaded into an Excel spreadsheet and the reduced χ^2 was calculated, assuming a function like Equation 1 but with a maximum of 5 exponential terms:

$$f(t) = A + \sum_{i=1}^5 B_i e^{-t/C_i} . \quad (2)$$

The reduced χ^2 is given by,

$$\chi^2 = \frac{1}{n - N} \sum_{i=1}^n \frac{(d_i - f(t_i))^2}{\sigma^2} , \quad (3)$$

where d_i represents the individual data points and t_i are the individual time values, n is the number of data points, N is the number of fit parameters, and σ is the estimated uncertainty in the individual data points. For each assay, σ was calculated as the standard deviation in the final 200 points.

The best values of A , B_i , and C_i were determined in stages. All of the B_i values were initially set to zero. Using the “Solver” routine provided in Excel, the χ^2 was minimized, first by allowing A , B_i , and C_i to vary and then reminimizing after freeing the parameters associated with each of the additional exponential terms in turn. In this way, very good fits were achieved. An example of the data, its fit, and the resulting residual is shown in Figure 1. The most interesting quantity extracted from the data, for this discussion, is the primary time constant. The primary time constant is defined as the C_i value corresponding to the largest magnitude B_i value for a given assay.

After extracting the primary time constants, the next step was estimating the uncertainties in the primary time constant values. This was done by repeating the minimization process with the primary time constant held fixed at various values. The reduced χ^2 increased as the primary time constant was changed from the optimum value. The high and low values of the primary time constant at which the reduced χ^2 was increased by 1.0 were noted. The assigned uncertainty was taken as half the distance between these high and low values. Briefly, this method estimates the uncertainty in a given parameter by using the change in the reduced χ^2 to define a confidence interval. More information can be found in the references.[4,5]

RESULTS

The results of the data analysis are provided in Table 1 and are plotted in Figure 2. Each group of points contains at least one assay with a source and one without a source. The differences between the points are of the order of the estimated uncertainties. The good agreement among points for each matrix supports the conclusion that the primary time constant extracted from the data reflects a physical property of the item that is independent of the power of the radioactive source. It is possible to distinguish among the different matrix materials simply by examining the primary time constants.

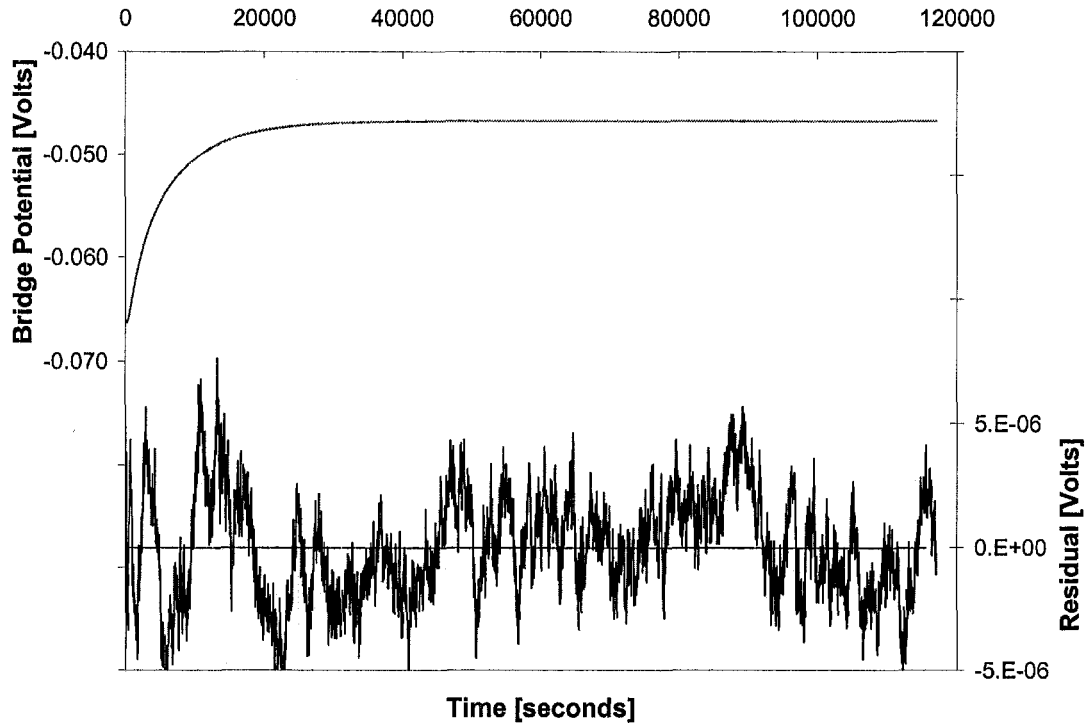


Figure 1: Calorimeter assay results for polyethylene beads without a source. The fit is indistinguishable from the data on the scale plotted. The residual reflects the intrinsic system noise.

The obvious question is what thermal property is driving the differences between the time constants. Table 2 summarizes the masses of the matrix materials, neglecting the mass of the source, the mass of the container, and the specific heat of each matrix. A strong correlation was found with the specific heat when the primary time constant was normalized by dividing by matrix mass. This is not surprising as specific heat is expected to play a role in heat transfer of the form $e^{-Dt/cp}$ where D is a constant, t is time, and cp is the specific heat. A plot of specific heat versus primary time constant divided by mass is shown in Figure 3. Also shown is a fit to the measured data points which yielded,

$$cp = 0.5476 \cdot \frac{C_1}{m}, \quad (4)$$

where cp is the specific heat in Joules per kilogram per degree Kelvin, C_1 is the primary time constant in seconds, and m is the mass in kilograms. Most of the scatter about the line can be attributed to uncertainties in the specific heat values which were obtained from standard references. Independent measurements of the specific heat for the materials used were not available. The values given are standard values for the materials used, however differences within categories of the material can be large depending on such factors as the alloyed composition, particle size, or processing techniques.

Not shown in Figure 3 are the data points from air and aluminum foil. In these cases, the primary time constant corresponds to the specific heat making the largest contribution to

the measurement time of the item. For most of the items measured, this is primarily the matrix material, as that is the most massive component. However, for air and aluminum foil, the mass of the matrix is small compared to the overall mass of the item, so it is unclear what mass should be used for normalization and what specific heat should be used for comparison.

Based on Equation 3, it is possible to estimate where items composed primarily of plutonium oxide and plutonium metal would be seen in Figure 3. The specific heat of plutonium oxide is 280 J/kg/K and of plutonium metal is 130 J/kg/K. The corresponding primary time constant divided by mass values are shown as squares in Figure 3. Clearly, for plutonium items of similar mass, it would be possible to distinguish between oxide and metal.

CONCLUSIONS

Using a relatively simple data-analysis technique it is possible to extract more than simply the power output from calorimeter data. Specifically, we have shown a strong correlation between the measured quantities of primary time constant and mass and the matrix material's specific heat. That the relationship exists is not surprising, however, the ability to extract the relationship from ordinary NDA-type measurements has not been previously seen.

As discussed previously, one application would be distinguishing between plutonium oxide and plutonium metal, a potentially valuable tool under various verification regimes. Further development of this technique should lead to additional applications. In principle, the other exponential terms contain information about the various components and materials making up the item. More detailed item information could be useful in establishing a unique fingerprint for the item or detecting item tampering.

ACKNOWLEDGMENTS

This work was funded by the Department of Energy, Office of Safeguards and Security.

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- [4] W.T. Eadie et al., Statistical Methods in Experimental Physics, North-Holland Publishing Company, New York, NY, 1971.

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Item Type	Reduced χ^2	Primary Time Constant (seconds)	Estimated Uncertainty in Primary Time Constant (seconds)
Air	1.34	1715.7	37.5
Air and source	2.72	1764.1	46.5
Aluminum	1.01	1924.8	17.0
Aluminum	1.73	1760.0	61.5
Aluminum and source	1.52	2027.4	26.0
Aluminum and source	1.59	1712.1	38.5
Aluminum bars	3.63	5319.8	15.5
Aluminum bars and source	3.54	5367.1	7.5
Copper	1.97	7878.0	41.0
Copper and source	2.19	7785.3	79.5
Lead	1.59	4403.3	17.0
Lead and source	0.65	4373.2	175.0
Poly	2.74	6981.0	130.0
Poly	1.62	7060.5	33.5
Poly and source	2.26	7117.3	55.0
Salt	2.88	5364.9	12.5
Salt and source	2.09	5388.2	16.5
Sand	1.49	6016.1	93.5
Sand	5.80	6028.5	15.0
Sand and source	2.58	6100.0	87.5
Steel	1.66	10450.7	32.5
Steel	1.47	10687.1	120.0

Table 1: Data Analysis Results. The primary time constant was determined by minimizing the reduced χ^2 . The uncertainty in the primary time constant was estimated by examining the change in χ^2 for different values of the primary time constant.

Matrix Type	Item Mass [kg]	Matrix Specific Heat [J/kg/K]
Aluminum	3.6	903
Copper	15.8	385
Lead	20.7	130
Steel	13.8	460
Polyethylene	1.7	2200
Salt	3.1	878
Sand	4.6	800
Air	0.0	?

Table 2: Matrix Properties. The approximate item mass and specific heat of the matrix material are summarized.

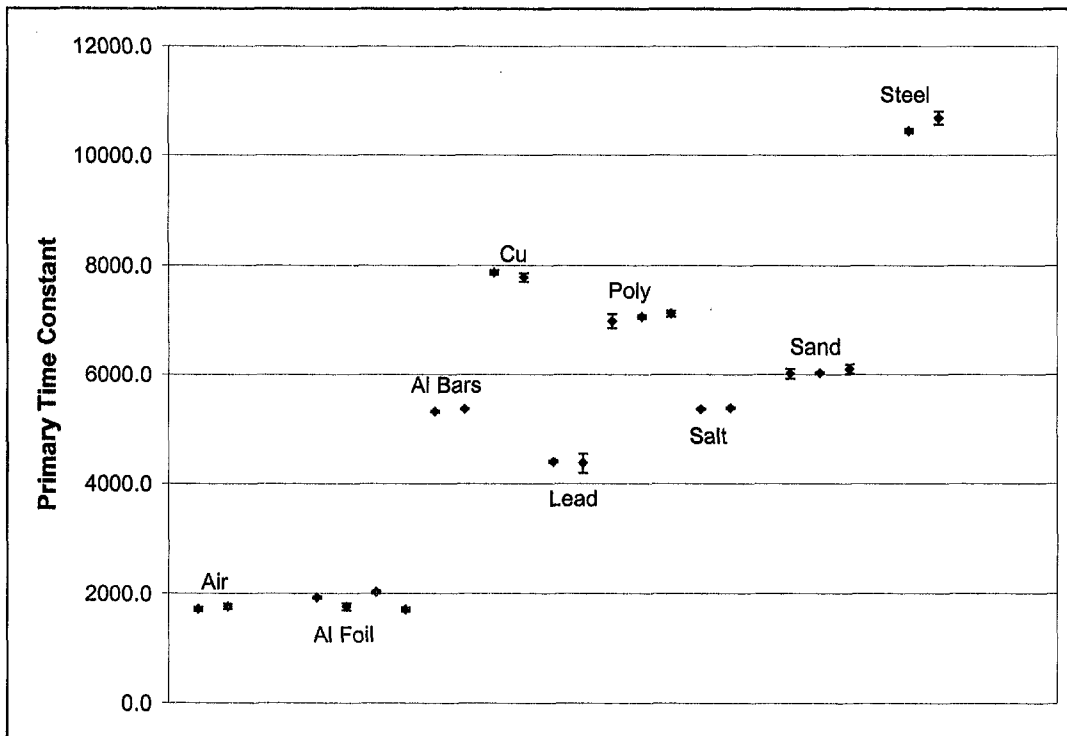


Figure 2: Primary time constants. The scatter between points corresponding to the same matrix is small.

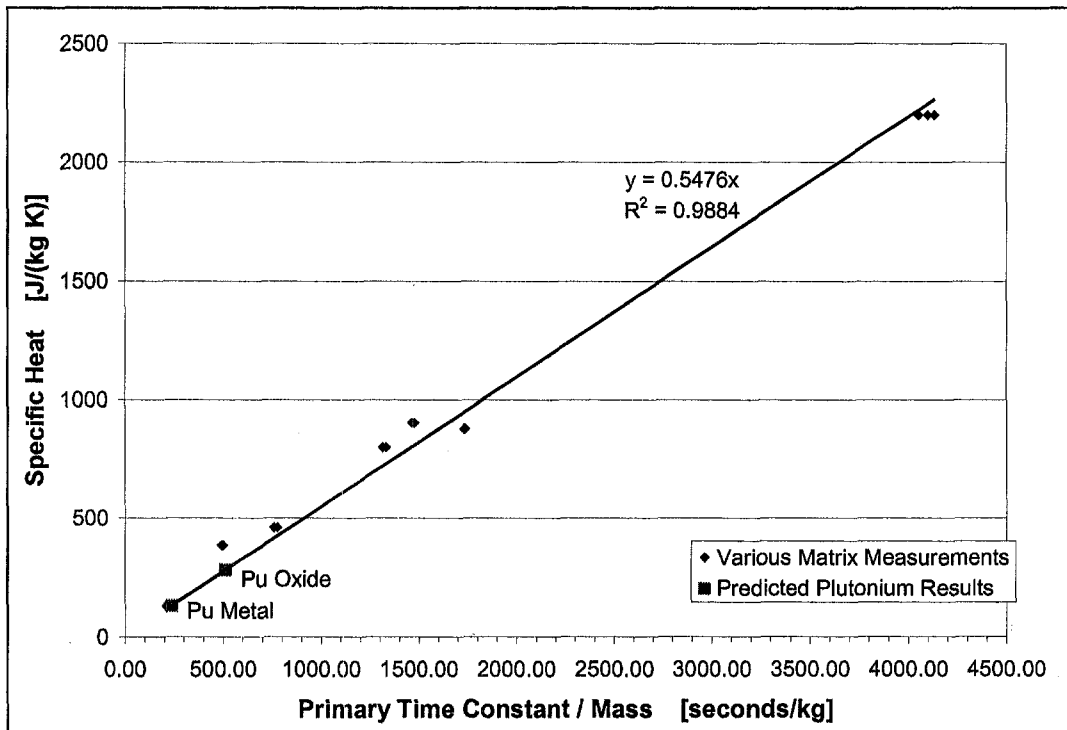


Figure 3: Specific heat correlation. The scatter about the fit may be attributed to uncertainties in the exact specific heat to use for the item.